



PATENT SPECIFICATION

NO DRAWINGS

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COMPLETE SPECIFICATION

Organopolysiloxane Compositions

We, GENERAL ELECTRIC COMPANY, a corporation organised and existing under the Laws of the State of New York, United States of America, residing at 1, River Road, Schenectady 5, New York, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to organopolysiloxane elastomers useful for making insulating tapes which are self - bonding over an extended period of time.

The employment of organopolysiloxane elastomers as electrical insulators has gained wide acceptance by industry. Insulating tapes in the form of semi - cured organopolysiloxane elastomers, or glass composites thereof, have been used extensively in a variety of electrical applications. Semi - cured tapes are desirable because they can be fused by use of heat and pressure after being applied. More recently, self - bonding, or auto - adhering organopolysiloxane tapes, such as taught in British patent 859,284, have become available to the art. These self - bonding tapes have an important advantage over semi - cured tapes. Self - bonding tapes can be directly wound onto an exposed electrical conductor, such as in a splicing operation, to form an integral electrical insulator without the use of the extreme conditions required when applying semi-cured tapes.

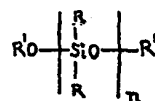
Although self - bonding tapes have important advantages over semi - cured tapes, experience has shown that after a relatively short shelf - period, such as a month or more, the ability of the tapes to self - bond is substantially diminished. Even if the tape is wrapped in a material such as cellophane, designed to protect materials from the effects of exposure to the atmosphere, only slight improvements in shelf stability are achieved. It would

[Price 4s. 6d.]

be desirable therefore to be able to provide the art with a self - bonding tape which can be advantageously employed in a variety of electrical insulating applications after an extended shelf - period.

It has been discovered that if certain organo - phosphorous - oxygen compounds, or mixtures thereof are utilized with a mixture of ingredients convertible to the elastomeric state comprising an organopolysiloxane polymer, a filler, and a boron - oxygen compound, the resulting composition can be formed into electrical insulating tapes which have self-bonding characteristics over extended periods of time, such as six months or more.

In accordance with the present invention there is provided an elastomer - forming composition comprising by weight (A) 100 parts of a polymer having the formula,



(1)

(B) 0.01 to 10 parts of an organo - phosphorous - oxygen compound selected from phosphonate esters, phosphonite esters, phosphite esters and phosphate esters, (C) 10 to 300 parts of a filler, and (D) .01 to 1 part of boron in the form of boron - oxygen compound, where n is an integer equal to from 100 to 10,000, each R is a monovalent hydrocarbon radical, a halogenated monovalent hydrocarbon radical, or a cyanoalkyl radical, and each R' is a hydrogen atom, an alkyl radical, or an $-\text{Si}(\text{R})_3$ radical.

Radicals included by R of formula (1) are aryl radicals, and halogenated aryl radicals, aralkyl radicals, haloaliphatic and cycloaliphatic radicals, cyanoalkyl radicals, Radicals

included by R^1 of formula (1) are hydrogen, methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, and octyl, R and R^1 separately can be all the same radical or any two or more of the aforementioned radicals respectively included above by either R or R^1 ; R is preferably methyl, or a mixture of methyl, phenyl, and vinyl, and R^1 is preferably $\text{Si}(\text{CH}_3)_3$.

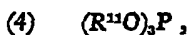
The organo - phosphorous - oxygen compounds that can be employed in the practice of the invention include esters of phosphonic acids,



esters of phosphonous acids,



phosphite esters,

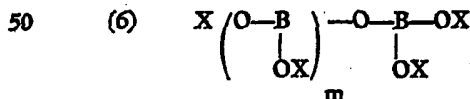


and phosphate esters,



where R^{11} is selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals included by R above, R^{111} is an alkyl radical included by R above, and a is 1 or 2. The preferred organo-phosphorous - oxygen compounds that can be utilized in the practice of the invention are the esters of phosphonic acids as shown in formula (2) e.g. dimethylmethylphosphonate, diethylethylphosphonate, diethylbutylphosphonate, dibutylethylphosphonate, dibutylbutylphosphonate, dipropylethylphosphonate, and dimethylphenylphosphonate. In addition, secondary esters of phosphonic acids are also operable. Examples of phosphonite esters according to formula (3) that are operable are dimethylmethylphosphonite and dibutylbutylphosphonite. Esters of phosphorous acid that can be utilized as shown in formula (4) include phosphite esters such as trimethylphosphite, triethylphosphite, tributylphosphite, or triphenylphosphite. Phosphate esters as shown by formula (5) include trimethylphosphate, triethylphosphate and triphenylphosphate.

The boron - oxygen compounds that are operable in the invention are preferably boric acid, H_3BO_3 , and boron oxides such as B_2O_3 , B_2O_2 , B_4O_7 . Boric acids and esters thereof that can be employed are included by the formula,



Cyclic organo borates,



are also operable, where m is 0, 1, 2 or 3, y is 3, 4 or 5, and X is a member selected from hydrogen and R^{111} radicals, where R^{111} radicals are as defined above.

Boron - oxygen compounds included by formula (6) and (7) are boric acids such as $\text{H}_3\text{B}_2\text{O}_4$, $\text{H}_2\text{B}_4\text{O}_7$, H_3BO_3 , $\text{H}_2\text{B}_2\text{O}_5$, HBO_2 , and esters of boric acid, e.g., alkyl borates aryl borates, substituted orthoborates, and cyclic organo borates.

The polymers of formula (1) can be chain-stopped with silanol radicals, alkoxy radicals, triorganosilyl radicals, or mixtures thereof. The polymers of formula (1) also can be utilized in the compositions of the present invention in the form of a mixture of polydiorganosiloxane polymers which are chain-stopped with silanol radicals, alkoxy radicals, and triorganosilyl radicals in which the organo radicals of such polymers are the same as the R radicals of formula (1).

The polymers of the present invention can be prepared by any one of several well known methods. For example, silanol chain - stopped polymers can be made by treating a highly viscous polydiorganosiloxane such as polydimethylsiloxane with water. Highly viscous polydiorganosiloxanes, i.e. polymers having a viscosity between 100,000 to 50,000,000 centipoises at 25°C ., can be made for example, by hydrolyzing diorganodichlorosiloxanes or mixtures of various diorganodichlorosilanes with water, and thereafter condensing the hydrolysis product with either acidic or alkaline catalysts such as hydrochloric acid, sulphuric acid and potassium hydroxide. Alternatively, one may heat cyclic polymers, for instance octamethylcyclotetrasiloxane, included by the formula,



(8)

where r is an integer equal to from 3 to 6, and R is as defined above, with an alkaline catalyst such as potassium hydroxide, or caesium hydroxide. Mixtures of various cyclic polymers can also be heated in a similar manner with an alkaline catalyst to effect the equilibration of the various diorganosiloxy units. The equilibration can be conducted in the presence of from about 0.001 to 0.1 per cent of alkaline catalyst based on the weight of the cyclic polymers of formula (8) or mixtures thereof at temperatures of from about 125°C . to 175°C . for times ranging from about 15 minutes to 2 hours or more. The alkaline catalyst in the resulting product can thereafter be neutralized with an aqueous mineral acid to yield a high molecular weight polydiorganosiloxane.

In order to obtain a silanol chain-stopped polymer as shown in formula (1), such as silanol chain - stopped polydimethylsiloxanes, or a silanol chain - stopped polymer composed of chemically combined dimethylsiloxo units, diphenylsiloxo units, and methylvinylsiloxo units, the aforementioned high molecular weight polydiorganosiloxane can be treated with water to reduce the viscosity of the polymer to a desirable range which can be between 2,000 to 50,000,000 centipoises at 25°C. This can be accomplished by blowing steam across the surface or through the high molecular weight polymer for a sufficient time to give the lower viscosity material having the desired silanol content. Such compositions and various methods for preparing the same are more particularly described in U. S. patent 2,607,792 and British patent 791,370.

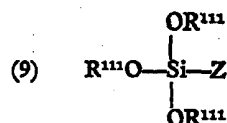
The use of steam in this fashion will cause a decrease in the viscosity of the polymer while at the same time the resulting linear polydiorganosiloxane will have terminal silicon-bonded hydroxy groups.

A method for making alkoxy chain-stopped polymers is to initially produce an intermediate halogen chain - stopped polydimethylsiloxane and to react this intermediate with an alcohol in the presence of a mild alkali. Another procedure involves equilibrating a cyclopolydiorganosiloxane such as octamethylcyclotetrasiloxane with a low molecular weight alkoxy chain - stopped polydiorganosiloxane. Methods for making $(R)_2SiO$ chain - stopped polymers are also well known such as by equilibrating a cyclopolydiorganosiloxane as shown by formula (8) with a hexaorganodisiloxane or decaorganotrisiloxane where the organo radicals are the same as the R radicals of formula (1). In addition, a hydrolyzate of a diorganodihalosilane or mixture thereof, can be equilibrated with triorganosiloxo units. Other methods are shown in E. G. Rochow, *An Introduction to the Chemistry of the Silicones*, 2nd Edition, New York, John Wiley and Sons (1951).

Various fillers, or mixtures thereof, can be employed to make the compositions of the inventions such as for example, titanium dioxide, lithopone, zinc oxide, zirconium silicate, silica aerogel, diatomaceous earth, calcium carbonate, fumed silica, precipitated silica, glass fibres, and iron oxide. The preferred filler is fumed silica because it is particularly useful in insulating applications. The amount of filler used can obviously be varied within wide limits, for instance, from about 10 to about 300 per cent by weight of filler based on the weight of the polydiorganosiloxane of formula (1). In instances where fumed silica is utilized, a proportion of from 20 to 50 parts, per 100 parts of polymer of formula (1) is preferred.

Various curing agents and mixtures thereof can also be utilized with the polydiorgano-

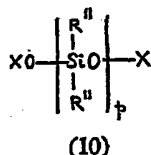
siloxane of formula (1) along with the filler and other ingredients to convert the resulting composition to the cured, solid, elastic state. Among such curing agents, there can be mentioned for example, benzoyl peroxide, tertiary butyl perbenzoate, bis - (2,4 - dichlorobenzoyl)peroxide, and dicumyl peroxide. These curing agents can be present in amounts ranging from about 0.1 to as high as 4 to 8 per cent, or more, by weight, based on the weight of the polymer. In instances where an alkyl borate is used as the boron - oxygen compound, in making the compositions of the invention, self - bonding compositions can be prepared that are curable at room temperatures. In addition to the polydiorganosiloxane of formula (1), filler, alkyl borate, and organo-phosphorous - oxygen compound, these room temperature curable materials may also contain organic silicates and certain metallic salts of alkyl carboxylic acids having from 8 to 20 carbon atoms in the carbon chain. For example, organic silicates in the form of an alkyl silicate (including a trialkoxyorganosilane), shown by the formula,



where R^{111} is as defined above, while Z is R^{111} or OR^{111} , or a halogenated product thereof, can be present in a range of from 0.1 to 10 per cent by weight of the polydiorganosiloxane of formula (1), while the metallic salt, for example, the octoate of zinc, tin, zirconium, lead antimony, or cadmium can be present as metal ion in a range of from 0.1 to 5 per cent by weight of the chain - stopped polydiorganosiloxane. High energy electron irradiation, or gamma irradiation, without the employment of a curing agent can also be employed to convert the various mixture of ingredients utilized in forming the self-bonding compositions of the present invention to the solid, elastomeric state.

In forming the self - bonding compositions of the present invention, the polymer, filler, and the organo - phosphorous - oxygen compound, can be mixed together in any desired manner. One procedure for example, is to add the filler to the polymer while the latter is being milled such as on a standard rubber mill. To the resulting mixture, or along with the filler there can be added suitable ingredients such as the boron - oxygen compound, the organo - phosphorous oxygen compound and various other ingredients such as structure additives and pigments. Preferably, the organo - phosphorous - oxygen compound is utilized in a proportion of from 0.1 to 1 part, per 100 parts of polymer. In terms of weight of boron, the boron - oxygen

compound is preferably utilized at from .07 to .7 part, per 100 parts of polymer. In addition, it also has been found expedient to utilize a chain - stopped oil,



where R^{II} and X are as defined above, p is an integer equal to from 3 to 70, inclusive, and preferably from 5 to 20, inclusive. In formula (10), R is preferably methyl, and X is preferably hydrogen or methyl. Where R^{II} is methyl in formula (10), and X is hydrogen, the oil can have a silanol content of by weight of from 2 to 15 per cent, and viscosity of 10 to 50 centipoises at 25°C.; it can be utilized in proportions of from 1 to 100, and preferably from 5 to 20 parts, per 100 parts of the polymer of formula (1). Where R^{II} and X are methyl in formula (1), the oil can have a methoxy content of from 2 to 30 weight per cent and a viscosity of from 5 centipoises or less to as high as 50 centipoises or more at 25°C. It also can be utilized at from 1 to 100 parts per 100 parts of polymer, while it is preferably used at from 5 to 20 parts per 100 parts of polymer. In addition to the aforementioned ingredients, heat stabilizers such as iron oxide, or aryl urethanes can be added in effective amounts such as in proportions of up to 4 parts of heat stabilizer per 100 parts of polymer.

Addition of the curing catalyst can be performed in any stage of the processing but it is often desirable to add it after mixing the polymer of formula (1) with the filler, boron-oxygen compound, etc.

The resulting curable composition then can be shaped by a conventional extrusion or calendering operation into tapes, including composites with glass cloth, or cloth containing fibers and synthetic fibers such as polyethylene terephthalate. Deposition of the curable composition onto a suitable cloth tape substrate can also be accomplished by dipping procedures, using a solution of the composition in a suitable organic solvent. Suitable organic solvents that can be employed are for example, toluene, benzene, and xylene.

The compositions of the present invention can be cured at a temperature within the range of 25°C. to 500°C. Although the time of cure will depend on such factors as the type of curing agent, concentration thereof and the temperature utilized, cure of the composition with a peroxide curing agent, for example, can be effected at a temperature between 315°C. to 500°C. for 1 second to 600

seconds. Excessively longer periods of cure at elevated temperatures have been found to adversely affect the self - bonding characteristics of the tape.

The following examples are given by way of illustration.

EXAMPLE 1

A silanol chain - stopped polydiorgano-siloxane composed of chemically combined dimethylsiloxo units, methylvinylsiloxo units and diphenylsiloxo units was prepared as follows.

There was equilibrated 100 parts by weight of octamethyl - cyclotetrasiloxane, 15 parts by weight of octaphenylcyclotetrasiloxane, and about $\frac{1}{2}$ part by weight of 1,3,5,7 - tetramethyl - 1,3,5,7 - tetravinyltetrasiloxane with about 0.1 per cent KOH catalyst and sufficient water to produce a polymer having a viscosity of about 20,000,000 centipoises at 25°C. To the resulting product there was added a stoichiometric amount of ammonium chloride in the form of an aqueous solution equal to the amount of KOH catalyst utilized in the equilibration reaction. The product was heated and then stripped of volatiles with steam.

There was added to 100 parts by weight of the above - described silanol - stopped polymer, 55 parts by weight of fumed silica, 15 parts by weight of a polydimethylsiloxane silanol chain - stopped oil having about 5 per cent silanol and a viscosity of about 20 centipoises at 25°C., 4 parts by weight of red iron oxide and 0.5 part by weight of boric acid, while the resulting mixture was being milled on a standard rubber mill. To the mixture there was then added $2\frac{1}{2}$ parts by weight of a mixture of 2,4 - dichlorobenzoyl peroxide in an equal weight of a low molecular weight polydimethylsiloxane oil.

There was added 0.135 part by weight of diethylethylphosphonate to 100 parts by weight of the above - described curable mixture, while it was milled. Tapes were prepared from the resulting curable composition by calendering it out to a sheet having a thickness of 20 to 30 mil, and curing the sheet for 15 to 30 seconds at 315°C. Tapes were also prepared following the same procedure from compositions free of the diethylethylphosphonate.

The tapes were then tested for ability to self - bond at room temperature after being exposed to the atmosphere for 1 day, 30 days, and 270 days by the following procedure:

A 10" long tape is wrapped under slight tension around a $\frac{1}{8}$ " diameter steel rod. After 24 hours the tape is checked to see if it can be unwound. If the tape self - bonds satisfactorily, it cannot be unwound without causing a cohesive tear. A cohesive tear indicates that the rubber has bonded integrally. If the tape does not self - bond, an adhesive tear results, that is, there is a clean separation between sections of the tape without substantial alteration of tape surface.

Table I below shows the self - bonding properties of the tapes in terms of the type of tear resulting after the stated period of exposure on the shelf. In Table I, "Alkylphosphonate" indicates diethylethylphosphonate.

TABLE I

Alkylphosphonate	Type of Tear		
	1 Day	30 Days	270 Days
Yes	Cohesive	Cohesive	Cohesive
No	Cohesive	Adhesive	Adhesive

EXAMPLE 2

A self - bonding composition was prepared following the procedure of Example 1, composed of 100 parts by weight of the same silanol - stopped polymer, 55 parts by weight of fumed silica treated with octamethylcyclotetrasiloxane, 15 parts by weight of a silanol-stopped polydimethylsiloxane oil having a viscosity of 20 to 30 centipoises, 4 parts by weight of iron oxide, $\frac{1}{2}$ part by weight of boric acid, and 0.25 part by weight of dibutylbutylphosphonate.

Tapes were prepared from this composition and tapes were also prepared by the same procedure free of dibutylbutylphosphonate. Some of the tapes were cured as in Example 1, while a few of the tapes were press - cured at

250°C. for 10 minutes. In addition, the press cured tapes were also heat - aged for 24 hours at 250°C.

The tapes that were cured by the method used in Example 1 were measured for self-bonding characteristics after 1 day, 30 days, and 270 days. A cohesive tear resulted after 270 days with the tapes made in accordance with the practice of the invention. The tapes free of dibutylbutylphosphonate did not self-bond satisfactorily when tested after 30 days exposure.

The press - cured tapes which also had been heat - aged for 24 hours at 250°C. were tested for Hardness "H", (Shore A), Tensile "T", (kg/cm²) and Elongation "E" (per cent) as shown in Table II.

TABLE II

Alkylphosphonate	Press Cured			Heat-Aged		
	10 Min. 250° C.			24 Hrs. 250° C.		
	H	T	E	H	T	E
Yes	40	67.5	800	60	68.2	660
No	45	70.3	800	70	38.7	400

The tapes made by the practice of the invention were also measured for dielectric strength, volume resistivity, power factor, etc. As shown in Table III, the tapes containing the dibutylbutylphosphonate were found to be as good in all respects as standard electrical insulating silicone tapes.

TABLE III

Electric Strength*	680
Dissipation Factor/60 CPS	.003
Dielectric Constant/60 CPS	3.5
Vol. Resistivity/Ohm/cm	4×10^{15}

*1" electrode under 10C oil, 0.5 Kilovolt/sec. rise

EXAMPLE 3

A room temperature curable self - bonding elastomer - forming composition is prepared by adding to 75 parts by weight of the silanol-stopped polymer of Ex. 1, 200 parts by weight of fine ground quartz, 25 parts by weight of the polydimethylsiloxane oil utilized in Example 1, 3 parts by weight of propyl silicate, 0.5 part by weight of lead octoate, and 0.5 part by weight of triethyl borate, and the resulting mixture is stirred in a doughmixer until a uniform composition is obtained.

There is added to 100 parts by weight of the above - described composition, while it is milled, 0.13 part by weight of diethylethyl-

phosphonate. Tapes are made from this composition by calendering procedures as in Example 1. Tapes are also made from the above composition free of diethylethylphosphonate. These tapes are allowed to cure at room temperature for 25 hours before they are measured for self - bonding. It is found that the self-bonding characteristics of the tape containing the diethylethylphosphonate remains substantially unchanged after 30 days shelf - period by the test procedure of Example 1. It is found that the tape free of diethylethylphosphonate does not self - bond after the 30 days shelf - period.

EXAMPLE 4

A mixture of 100 parts by weight of octamethylcyclotetrasiloxane, 0.23 part by weight of 1,3,5,7 - tetramethyl - 1,3,5,7 - tetravinyltetrasiloxane, 0.059 part by weight of decamethyltrisiloxane and 0.001 part by weight of KOH was equilibrated at a temperature between 150°C. to 195°C. until a polymer was produced having a viscosity of about 20 million centipoises at 25°C. This polymer was composed of chemically combined dimethylsiloxy units and methylvinylsiloxy units, and chain - stopped with trimethylsiloxy units.

There was added to 100 parts by weight of the above - described trimethylsiloxy-stopped polymer, 50 parts by weight of fumed silica, 12 parts by weight of a methoxy chain-stopped polydimethylsiloxane oil having a viscosity of about 15 centipoises, 4 parts by weight of red iron oxide, 7 parts by weight of triethyl borate, and 0.6 part by weight of dibutylbutylphosphonate, while the resulting mixture was being milled on a standard rubber mill. To the resulting milled mixture there was then added 3 parts by weight of a mixture composed of equal proportions by weight of dicumyl peroxide and a mixture of 2,4 - dichlorobenzoyl peroxide in an equal weight of a low molecular weight polydimethylsiloxane oil.

Tapes were prepared from the resulting composition by calendering it to a sheet having a thickness of about 20 to 30 mills in accordance with the procedure of Example 1. Tapes were also made following the above procedure except that no dibutylbutylphosphonate was utilized.

The tapes were then tested for self - bonding after exposure to the atmosphere at room temperature, for 1 day, 30 days and 270 days as shown in Example 1. It was found that the tapes made by the practice of the invention exhibited a cohesive tear after 270 days exposure, while the tapes free of dibutylbutylphosphonate failed in less than 30 days. In addition to the above tests, the tapes containing the dibutylbutylphosphonate were also found to exhibit substantially the same electrical properties as shown above in Table III, and substantially the same physical properties as shown in Table II, after a press-cure at 150°C.

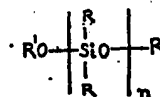
for 10 minutes, and a post - cure for 24 hours at 250°C.

As shown in the examples and in the Tables, the self - bonding tape compositions of the present invention possess unique and valuable properties. As a result of the present invention, there is provided superior electrical insulating tapes which can be utilized after extended shelf - periods in a variety of electrical insulating applications.

WHAT WE CLAIM IS:—

1. An elastomer - forming composition comprising by weight:

(a) 100 parts of a polymer having the formula:



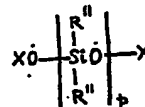
where n is an integer equal to from 100 to 10,000 inclusive, each R is a monovalent hydrocarbon radical, a halogenated monovalent hydrocarbon radical, or a cyanoalkyl radical, and each R^1 is a hydrogen atom, an alkyl radical or a $-\text{Si}(\text{R})_3$ radical;

(b) 0.01 to 10 parts of an organo - phosphorus - oxygen compound selected from phosphonate esters, phosphonite esters, phosphite esters and phosphate esters,

(c) 10 to 300 parts of a filler, and

(d) 0.01 to 1 part of boron in the form of a boron - oxygen compound.

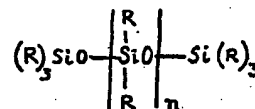
2. A composition as claimed in Claim 1, wherein there is added 1 to 100 parts by weight of a polydiorganosiloxane oil having the formula,



where p is an integer equal to from 3 to 70, inclusive, each R'' is a monovalent hydrocarbon radical or a halogenated monovalent hydrocarbon radical, and each X is hydrogen or an alkyl radical.

3. A composition as claimed in claim 2, wherein there is added 5 to 20 parts by weight of said polydiorganosiloxane oil.

4. A composition as claimed in any preceding Claim, wherein the polymer has the formula:

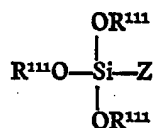


where R and n are as defined in Claim 1.

5. A composition as claimed in Claim 4, wherein the polymer employed is a trimethyl-

siloxo chain - stopped polydiorganosiloxane composed of chemically combined dimethyl siloxy units, methylvinylsiloxo units and diphenylsiloxo units.

- 5 6. A composition as claimed in any preceding Claim, wherein there is added from 0.1 to 10 parts by weight of a compound of the formula:



- 10 where R^{111} is an alkyl radical, and Z is R^{111} , OR^{111} , or a halogenated alkyl radical.

7. A composition as claimed in any preceding in Claim, wherein the organo - phosphorus - oxygen compound is a dialkylalkylphosphonate, such as a dibutylbutylphosphonate, or a diethylethylphosphonate.
- 15

8. A composition as claimed in any preceding Claim, wherein there is employed as the boron - oxygen compound, boric acid or triethylborate.

9. A composition as claimed in any preceding Claim, wherein fumed silica is employed as the filler.

10. A tape made from the composition claimed in any of the preceding claims.

11. The composition in accordance with claim 1 substantially as described herein in any one of the foregoing Examples.

12. A tape made from a composition in accordance with Claim 1 substantially as described herein in any one of the foregoing Examples.

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